ISOTOPICALLY HEAVY NITROGEN IN THE BENCUBBIN BRECCIA: A LONELY ANOMALY. G.P. Rooke¹, I.A. Franchi¹, A.B. Verchovsky¹, M.M. Grady^{1,2} and C.T. Pillinger¹ ¹Planetary Sciences Research Institute, Open University, Milton Keynes, UK. ²Mineralogy Dept., The Natural History Museum, Cromwell Road, UK.

Bencubbin, a stony iron brecciated meteorite, consisting of metal clasts, silicate clasts and shock melted mixture of metal and glass, is almost unique in having a whole rock ¹⁵N of nearly +1000‰ [1]. Earlier stepped heating experiments on acid residues of the meteorite showed that the vast majority of this heavy N is concentrated in a carrier phase, which once the metal and most of the glassy matrix was removed, combusted at a relatively low temperature of 400°C [2]. However, despite the labile nature of this component, these early studies were unable to properly constrain the site and origin of the heavy N.

More recently, the original 6M HCl residues were reinvestigated, but this time with conjoint analysis of ⁴He, ¹²C, ^{20,21,22}Ne and ^{36,38,40}Ar together with the nitrogen [3], in the hope that any distinctive noble gas signatures would help constrain the origin of the N. The result of that study was that no major noble gas component was identified which could be directly associated with the sharp release of N at 400°C. However, a small component of Ar was released at 300°C. To link this Ar with the heavy N, it was suggested that the carrier of the N had been acid or radiation damaged. Given that the most probable host for the heavy N was a Cr-rich sulphide [2], it is entirely possible that this phase may have indeed suffered some damage during treatment with 6M HCl.

To minimise the damage to the N carrier a milder dissolution treatment has been performed on a new sample of Bencubbin using CuCl₂ solution. Using this reagent only the Fe-Ni metal clasts and metal in the matrix should be removed, leaving the sulphides, silicate clasts and silicate glass in the matrix virtually untouched. A 0.85g sample of Bencubbin, a mixture of mostly matrix with some metal and silicate clasts, was dissolved in 1M CuCl₂ in an ultrasonic tank for 23 hours, the ultrasound ensuring that the Cu precipitated from the dissolution reaction remained fine grained. This Cu is then readily redissolved and removed by a number of quick washes in 0.5M HCl. The resulting residue was composed of a powder (2.7 wt% of the whole rock) and 3 "clasts" (27.9 wt% of the whole rock). These "clasts" were severely "honey-combed", but still coherent blocks, indicating that most of the silicate glass portion of the matrix was unaffected by the CuCl₂ treatment and continued to bind the rock together, with only the metal portion of the matrix, along with the metal clasts having been removed. The 6M HCl treatment on the other hand resulted in a far greater weight loss and left essentially only a fine powder [2].

As was found for the 6M HCl residue there is a concentration of N in the residue, with stepped combustion experiments showing that the nitrogen is more concentrated in the powder (11 ppm whole rock) than in the clasts (7 ppm whole rock). However, combined, these concentrations are 3x lower than that in the 6M HCl residue (58 ppm whole rock). This discrepancy is difficult to explain as the CuCl₂ treatment was expected to be far less harsh than the 6M HCl. However, the complex, brecciated nature of this meteorite means that sample heterogeneity is a more than plausible candidate to account for the lower concentration in the CuCl₂ residue. The N yield from the "clasts" appears to reveal a minor peak of heavy N at 400°C, superimposed on a release of low temperature contamination, and a major peak starting at 1050°C coinciding with a major release of planetary Ar (³⁶Ar 2400x10⁻⁸ ccg⁻¹ residue), and spallogenic Ne (²¹Ne 10x10⁻⁸ ccg⁻¹ residue). The major release is identical to that displayed by the whole rock showing that much of the carrier is still shielded by a silicate component.

The release of nitrogen from the powder is virtually identical to that from the 6M HCl residue and coincides with the minor release from the "clasts". Unlike the "clasts" residue, however there was no release of Ar or Ne, above blank below 500°C. Equally there were no releases over 1050°C (see figure).

Further spallogenic Ne and planetary Ar releases were detected between 600°C and 1000°C together with a possible planetary Ne peak at 650°C, but no nitrogen was found to coincide with these releases. Total ³⁶Ar (600x10⁻⁸ ccg⁻¹ whole rock) and Ne (²¹Ne 3.5x10⁻⁸ ccg⁻¹ whole rock, ²⁰Ne 11.1x10⁻⁸ ccg⁻¹ whole rock) concentrations, calculated using both samples, are in agreement with previous single or 4 step noble gas analyses [5,6]. In contrast the concentration of ³⁶Ar in the 6M HCl residue was much lower than in either of these samples (3x10⁻⁸ ccg⁻¹ whole rock) with virtually no measurable Ne [4]. This shows that the bulk of the noble gases released between 600°C and

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 1000° C in the $CuCl_2$ residues resides in the silicate clasts or glassy matrix as these were destroyed or not present in the 6M HCl residue. The small release at 300° C in the 6M HCl residue, but not seen in the $CuCl_2$ residues must therefore have originated from acid damaged remains of the silicate host material and is not linked to the heavy N carrier.

The CuCl₂ treatment has preferentially released some of the heavy N carrier from the original sample where it was trapped in a high temperature phase. This phase appears to have included Ne and Ar but these are not intimately linked to the heavy nitrogen carrier. This disassociation of noble gases and the heavy N in Bencubbin although at first appearing a negative result in helping determine the origin of the anomaly may in fact help still constrain its history.

Previous suggestions about the origin of this ¹⁵N enrichment have included nucleosynthetic processes, extreme isotopic fractionation and spallogenic reactions [1,2]. A spallogenic origin would also be expected to produce a parallel signature in Ne and Ar but this is not observed, thereby ruling out this possibility. Equally, all stellar condensate grain populations found to date contain a number of elements with isotopic compositions which are clearly non-solar, whereas no such comparable signatures have been found in any of the Bencubbin residues for some of the mostly likely elements - C, Ne, S and Ar. Is the N anomaly in Bencubbin in itself anomalous in having no apparent supporting anomalous signatures? This suggests that we may be dealing with something all together different from the stellar condensates.

Having ruled out spallation and nucleosynthesis this leaves some extreme isotopic fractionation as the most viable type of process to account for the ¹⁵N enrichment. It is difficult to constrain the exact mechanism currently but some type of ion molecule reaction would be a good candidate, most probably in part of the dense molecular cloud which collapsed to form the solar nebula. Enrichments in ¹⁵N up to a factor of 2.5 have been predicted for such reactions at low temperatures (<20K) involving only H [7]. Fixation of the enriched N into a condensable species and thereafter into ice/dust grains could then proceed with elements closer to their final solar nebula isotopic compositions.

References

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